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THE RESPONSE OF PREMIXED FLAMES TO PRESSURE OSCILLATIONS

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Abstract

Recent measurements of the direct response of premixed flames to acoustic pressure fluctuations have shed doubt on the validity of analytical models that use irreversible one-step chemistry [1], and suggest that more realistic chemical kinetic models are needed to fully describe the unsteady dynamics of premixed flames. However, in the analysis of the experimental results some doubts subsisted concerning the exact relation between the intensity of emission from the excited OH* radical, used to determine the flame response, and the unsteady reaction rate given by the theoretical analyses. Combining experimental and numerical approaches on premixed methane-air flames, we propose corrections to give more confidence in the experimental results and to confirm the need for further investigations on the dynamics of unsteady premixed flames.

INTRODUCTION

It is well known that strong thermo-acoustic instabilities can be driven by fluctuations of flame surface area, induced either by convective effects [2,3] or by the effect of acoustic accelerations [4,5], or by system instabilities resulting, for example, from a coupling between the combustion chamber and the feed-line of the burner [6,7]. However the primary amplification of acoustic waves can also result from the direct effect of the local pressure on the chemical reaction rate [8]. The low frequency response of flame chemistry to acoustic pressure is always in phase and is thus always destabilizing when acoustic damping is neglected.

The steady-state response of a flame is fairly weak, but analytical studies predict that the unsteady part of the mass flux through the flame should increase with frequency [8,9,10]. It is thus interesting to experimentally investigate the unsteady response of flames to acoustic pressure oscillations and compare the experimental results with the response predicted by the theoretical analyses. Such results are also relevant for the prediction of the unsteady response of premixed flamelets in the context of a theory for turbulent flames, or in view of describing flame extinction by rapid pressure fluctuations [11,12,13].

In order to investigate the response of premixed flames to pure pressure oscillations, we have used the chemiluminescence of OH* radicals as a marker of the reaction rate and we have used a technique of parametric restabilisation [14] to obtain a perfectly flat flame in order to avoid the effect of flame area variations. Previous work [1] has shown that experimental measurements were in better agreement with the theoretical reduced heat release rate fluctuations than with the calculated mass consumption rate fluctuations, both for the amplitude of the flame response and for the phase of response with respect to the pressure fluctuations. An attempt has been made to improve this agreement by considering a more realistic model for the chemical kinetics [15], see fig.1. However there are still some questions concerning the exact relation between the chemiluminescence of OH* radicals and the heat release rate.

After a brief description of the theoretical background and of the experimental set-up, we will compare experimental and numerical results on flame speed and OH* emission. This will lead us to introduce correcting factors on both the theoretical and experimental results.

THEORETICAL BACKGROUND

Combustion reactions are governed by an Arrhenius type law, giving a high sensitivity of the reaction rate to both pressure and temperature variations. Using asymptotic methods with a large dimensionless activation energy, or Zeldovich number, β :

$$\beta = \frac{E_A(T_b - T_u)}{RT_b^2} \gg 1, \quad (1)$$

where E_A is the activation energy, T_b and T_u are the temperatures of the burnt and unburnt gas respectively, and in the distinguished limit $\beta(Le - 1) \approx 1$ where $Le \equiv D_{th}/D_{mol}$ is the Lewis number of the deficient reactant in the mixture, various authors have evaluated the unsteady part of the mass consumption rate in response to acoustic pressure fluctuations [8,9]. They consider the frequency domain where the acoustic period, $1/f$, is of the order of or shorter than the flame transit time τ_f . If the acoustic time is not too much smaller than the transit time, the flame thickness is negligible compared to the acoustic wavelength and acoustic pressure gradients within the flame zone can be neglected.

The reaction rate of premixed flames is sensitive to both the density and the temperature of the reactants. Acoustic waves are adiabatic, so pressure oscillations are accompanied by temperature oscillations of the same order of magnitude:

$$\frac{\delta T}{T_u} = \frac{\gamma - 1}{\gamma} \frac{\delta p}{P_0}, \quad (2)$$

where γ is the ratio of specific heats. It is easy to see that for an Arrhenius reaction law with a large Zeldovich number, the reaction rate is β times more sensitive to a temperature fluctuation than to a pressure fluctuation of the same relative amplitude. The authors of [8,9] have retained only the response of the reaction rate to temperature oscillations in the acoustic wave and have calculated the unsteady response of the mass flux through the flame. The authors of [8] have also given an expression to calculate the unsteady response of the heat release rate. Considering only the unsteady response of the heat release rate, which is the important parameter for thermo-acoustic instability, the results of [8] can be written [1]:

$$\begin{aligned} \frac{\dot{Q}'/\bar{\dot{Q}}}{p'/\rho c^2} &= \frac{E_A}{RT_b} (\gamma - 1) \frac{(q - 1)}{2} \frac{A(\omega)}{B(\omega)} \\ &\text{with} \\ A(\omega) &= \{q - (T_b - T_u)/T_b\} q \\ B(\omega) &= (q - 1)q^2 - \frac{\beta}{2}(Le - 1)(1 - q + 2i\omega\tau_f) \\ q(\omega) &= (1 + 4i\omega\tau_f)^{1/2} \end{aligned} \quad (3)$$

where $\bar{\dot{Q}}$ is the mean heat release rate, γ is the ratio of specific heats, p is the pressure, ρ is the mass density, c is the sound speed, ω the pulsation and the prime denotes the fluctuating part of a quantity. The calculated phase response tends towards zero at infinitely low and high frequencies, and has a maximum phase lag of approximately $\pi/4$ when the acoustic period is of the order of the flame transit time. Figure 1 shows our experimental measurements of the amplitude and phase response of

chemiluminescence of lean methane-air flames to pressure oscillations. Both the amplitude and phase of response tend to be above the analytical predictions for the response of the heat release rate.

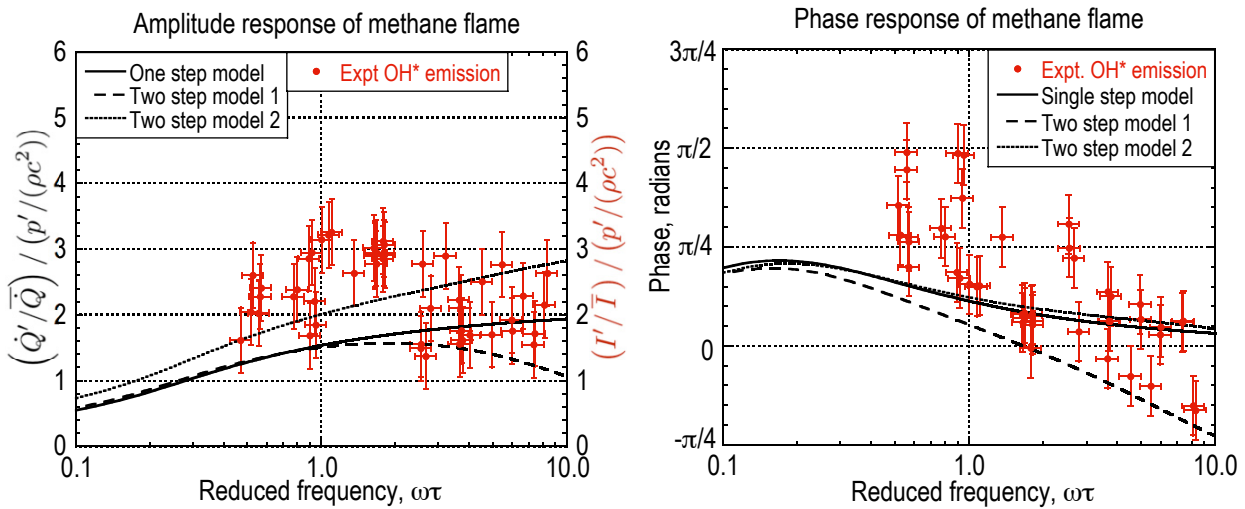
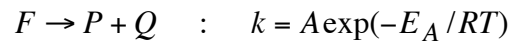
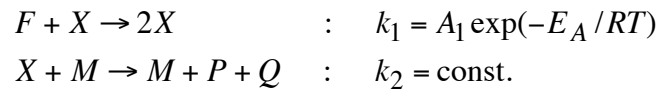


Figure 1: Comparison between the reduced reaction rate fluctuations measured by spontaneous light emission of lean methane-air flames, [1], and heat release rate fluctuations calculated by asymptotic methods for a unit Lewis number with three different chemical reaction schemes [15].

The experimental measurements are compared to the analytical calculations of the heat release rate using three different kinetic models [15]: a single one-step irreversible reaction governed by an Arrhenius law with a large activation energy:



and a two-step model of reaction kinetics consisting of an irreversible chain-branching step with a large energy of activation followed by an exothermic chain-breaking reaction whose reaction rate is not sensitive to the temperature:



The second exothermic reaction can either be frozen in the preheat zone (Model II) or can be allowed to take place anywhere that X is present (Model I). F represents the fuel, P the reaction products and Q the heat release. X represents the intermediate radical(s) and M is a third body. The formulation of the chain breaking reaction makes the reaction first-order and greatly simplifies the analytical calculations. It can be seen that the experimental and analytical results have the same order of magnitude. However one may still question the relevance of OH^* chemiluminescence as a linear measure of the reaction rate of premixed flames subjected to pressure oscillations. The global heat release rate and the total OH^* emission intensity obviously both increase linearly with flame surface area. However both quantities are also functions of:

- the equivalence ratio
- the temperature of the unburnt gas
- the pressure

The objective of this paper is to investigate the relation between OH^* emission and reaction rate when these parameters are varied, using both experimental and numerical tools, and then to use the results to correct the experimental data for unsteady measurements of OH^* emission.

EXPERIMENTAL APPARATUS

A premixed flame was stabilized in a cylindrical burner composed of a half-open Pyrex tube fed with the combustible mixture at the closed end. A porous plate made of sintered brass was placed just above the gas entry to laminarise the flow. The reactants, methane or propane and air, were regulated using sonic nozzles and the combustible mixture was produced in excess in order to permit continuous adjustment of the flow rate in the main burner without any risk of changing the composition. The excess mixture was consumed in a secondary burner. Standing acoustic modes of the tube were excited using a loudspeaker situated at the closed end.

A detailed description of the apparatus, and the method for monitoring the amplitude and phase of the acoustic field at the flame front are given in [1]. The flame is maintained perfectly planar by parametric restabilization [5,14]. This is accomplished by placing the flame close to the velocity anti-node of a low frequency standing acoustic wave. For certain values of the periodic acceleration imposed by the acoustic velocity field it is possible to obtain a perfectly planar flame. Figure 2 shows a flat flame obtained by parametric restabilization and maintained at a fixed position in the main burner by careful adjustment of the flow rate.

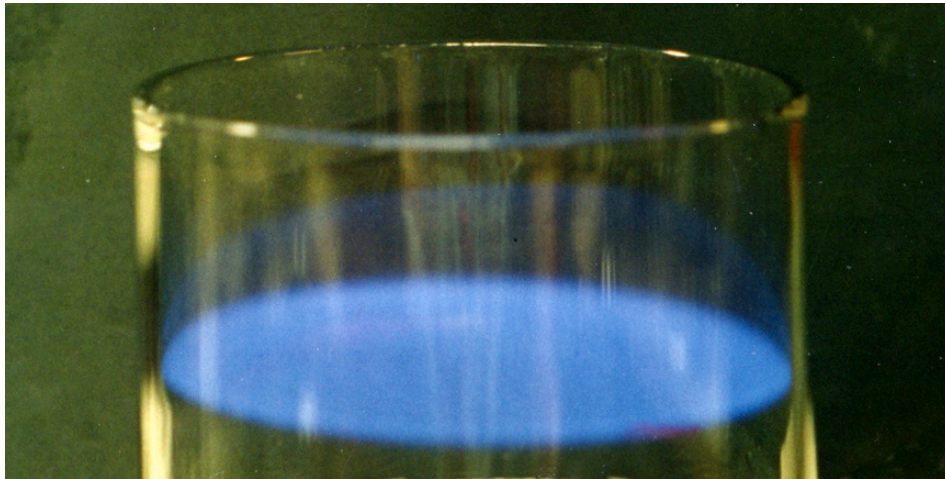


Figure 2 : Planar methane-air flame, equivalence ratio 0.68, stabilized in a burner 10 cm diameter.

The flame is then perturbed using the pressure anti-node of a *second* acoustic field at a higher frequency and we monitor the unsteady response of the flame to this second acoustic field. The non-dimensional frequency, $\omega\tau_f$, can be varied by changing either the laminar flame speed (τ_f), or by changing the frequency of excitation using resonances of higher harmonics and also by using tubes of different length ($0.6 < L < 3\text{m}$).

The OH* chemiluminescence originating from the central part of the flame was collected via a UV mirror placed above the burner exit. It was then imaged onto a photomultiplier tube through a quartz lens and an interference filter centred at 307nm with a band pass of 16nm. The whole optical system could be translated vertically to maintain a constant distance between the flame front and the optical system. The gain of the photomultiplier tube was also maintained at a fixed value. In this way, the DC part of the photomultiplier signal can be related to the mean OH* emission per unit surface of the flame. The AC component of the photomultiplier signal is related to the response of the flame to acoustic pressure oscillations at the reaction zone.

RESULTS

Effect of equivalence ratio and temperature on laminar flame speed and intensity of OH* emission

In order to investigate the relation between the mean intensity of OH* emission and the mass consumption rate, the flame velocity was measured using 1-D laser velocimetry in the presence of acoustic restabilisation. The flame was initially placed 1 cm downstream the point of measurement and allowed to drift slowly upstream (less than 1 cm/min). A typical record of the axial gas velocity is given in fig.3a showing the superposition of the mean flow velocity (here $\approx 0.15\text{m/s}$) and the acoustic velocity, whose amplitude is typically 4 to 5 times the laminar flame velocity (here $\approx 0.75\text{m/s}$), so that the flow direction is periodically reversed. Since the acoustic forcing is perfectly monochromatic, the gas velocity can be locally fitted with the function $u(x) = u_g(x) + u_{ac} \cos(\omega t + \varphi)$ to extract the mean gas velocity $u_g(x)$. The flow profile upstream of the flame front is not quite flat, but must become flat at the planar flame front, the flame thus induces a small negative velocity gradient on the axis. The laminar flame velocity was evaluated by extrapolating the gas velocity to the supposed abscissa of the reactive layer, estimated to be the point at which oil droplets disappeared (fig.3b). The position of the reactive layer could be evaluated with an error of about 0.5 mm. Since the gradient of the flow was very small, $< 4 \text{ s}^{-1}$, the uncertainty in the measured flame speed was less than 0.2 cm/s.

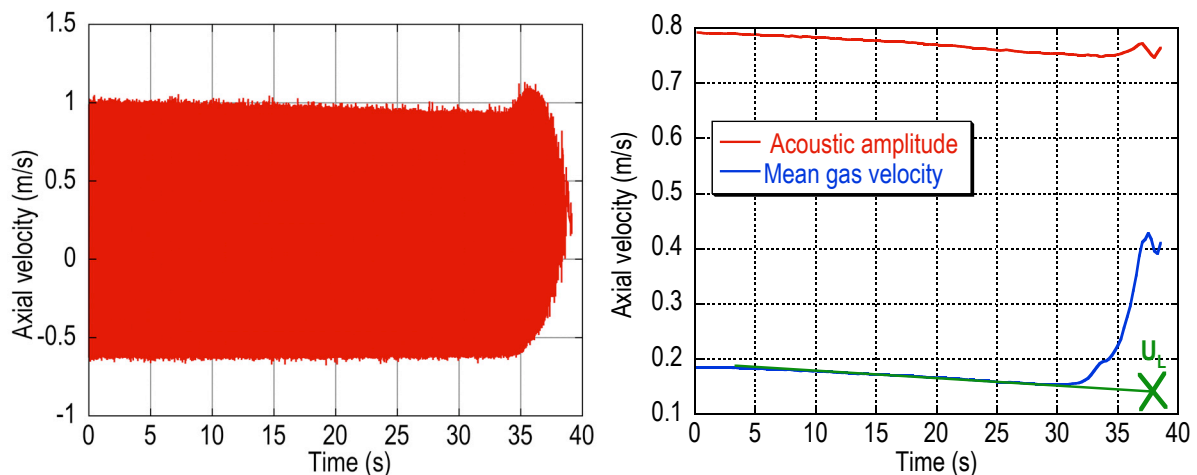


Figure 3 : Measurement of laminar flame speed in the presence of acoustic restabilisation.

Left: raw velocity just upstream of the flame. Right: analysed velocity field.

Methane flame, equivalence ratio=0.68 and gas temperature=36°C.

Because of the constraints of parametric restabilisation, the measurements of flame speed were limited to a narrow range of equivalence ratios between 0.6 and 0.7. The temperature of the unburnt gas was varied from 22°C to 60°C (fig.4). Our measured flame velocities compare well with the recent measurements of Bosschaart and de Goey [16] if we suppose that their fresh gas temperature was 20°C, and indicate that previous measurements, such as those of Yamaoka and Tsuji [17], were slightly overestimated.

The intensity of OH* emission was measured in the same conditions (fig.5) and compared to the results of steady-state numerical simulations using two different concurrent kinetic mechanisms to describe production of the excited OH* radical in the reaction zone [18,19].

As a basis kinetic scheme we have used the reaction mechanisms proposed by A.A Konnov [20] with 127 species and 1098 reactions. This set of reaction mechanisms provided laminar flame speeds in reasonable agreement with our experimental measurements, and in much better agreement than the

commonly used GRI-Mech3 scheme [21] that gave laminar flame speeds (not shown here) about 5cm/s too high for the range investigated.

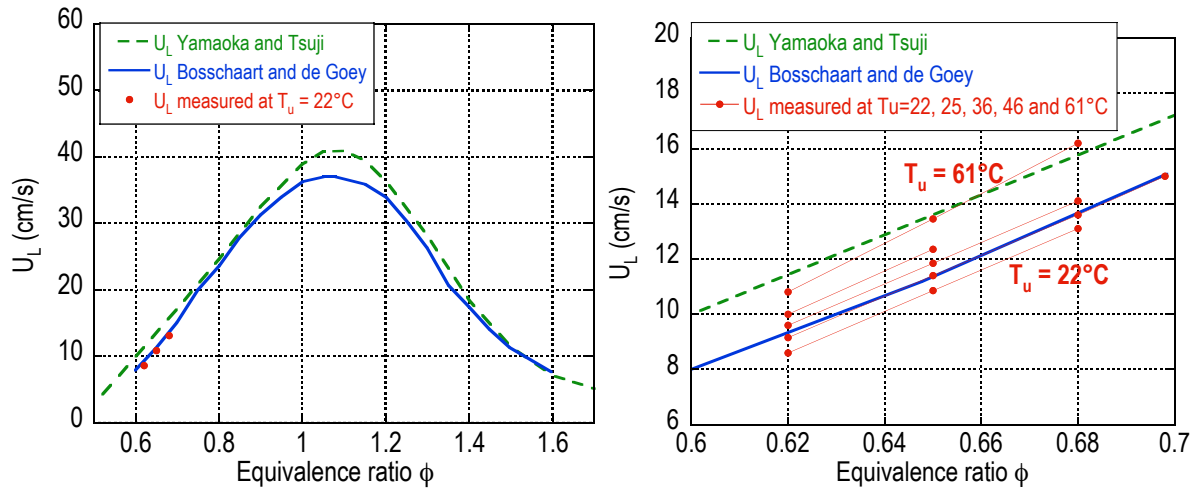
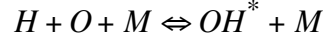
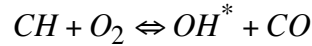


Figure 4. Comparison between our laminar flame speed measurements on a planar flame and measurements reported in references [16] and [17].

The excited OH^* radical is not part of either of the above schemes. We have grafted and compared two different kinetic schemes for OH^* production and de-excitation. The first mechanism, proposed by L.C.Haber [18], assumes that the radical OH^* is produced via $\text{HCO} + \text{O} \rightleftharpoons \text{OH}^* + \text{CO}$ while the second, proposed by Hall and Petersen [19], uses a more classical path for OH^* production:



The rate of photon emission per unit volume is then given by

$$\frac{dN_v}{dt} = k_j T^{n_j} e^{-E_{A_j}/RT} [\text{OH}^*] N_A \quad (4)$$

where $[\text{OH}^*]$ is the molar concentration of the excited radical, N_A is Avogadro's constant, and k_j , n_j , and E_{A_j} are the rate constants for OH^* de-excitation by photon emission [18,19]. The total number of photons per unit time and per unit surface of the flame was simply obtained by numerical integration through the flame.

In our measurements it was not possible to obtain an absolute calibration of luminous intensity with good precision. We thus choose to rescale the results to make the experimental and numerical values coincide at a reference point near the centre of our range of parameters: $\phi=0.65$ and $T_u=25^\circ\text{C}$. It can be seen from fig.5 that the OH^* mechanism of Konnov+Hall&Peterson (red curves) reproduces our experimental results more closely than the mechanism of Konnov+Haber (dotted blue curves), both for the variation with equivalence ratio and for the variation with temperature. Moreover the mechanism of Konnov+Haber predicts OH^* intensities 745 times lower than Konnov+Hall&Peterson. This is a large difference and our estimation of the absolute intensity was also in better agreement with the Konnov+Hall&Peterson scheme. We thus retain this latter scheme for the comparisons below.

Effect of pressure on OH^* emission

It is known that non radiative de-excitation of the OH^* radical through collisions (quenching) is faster than radiative de-excitation and increases with increasing pressure, leading to a negative effect of pressure on OH^* chemiluminescence. Acoustic pressure fluctuations will decrease OH^* intensity through pressure quenching and increase OH^* intensity through the effect of pressure and temperature

on reaction rate. In order to investigate the effect of an acoustic pressure oscillation on OH* emission, we have calculated OH* emission intensity using the scheme of Konnov+Hall&Peterson for both an isothermal compression and also for a set of pressures and temperatures where $P^{1-\gamma}T^\gamma$ is maintained constant (adiabatic compression). This calculation is steady-state, (or zero-frequency).

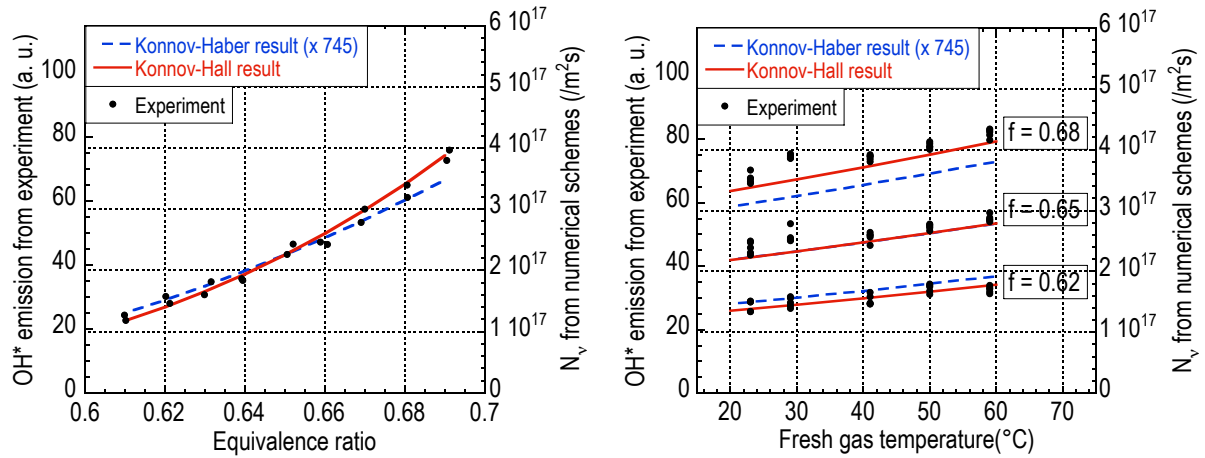


Figure 5. Comparison between experiment and numerical evaluations of OH* emission as a function of equivalence ratio (left) and fresh gas temperature (right) for two different reaction schemes.

These numerical results are shown in figure 6a. It can be seen that the isolated effect of pressure (blue curve) causes the OH* intensity to decrease, as expected. The effect of temperature increase during an adiabatic compression (red curve) counteracts the effect of pressure, however the slope of the curve is still negative. This behaviour would lead to a chemiluminescence signal whose phase is in opposition to the acoustic pressure. This is contrary to our experimental observations, see fig.1 right.

In view of the lack of consensus for the chemical kinetics of OH* and the possible uncertainty in the pressure response of the Hall&Peterson mechanism used above, we have searched the literature for relevant experimental data. Higgins et al. [22] have measured the effect of static pressure (but not temperature) on OH* chemiluminescence from a confined conical flame. They find the following empirical correlation between the intensity, I , of OH* chemiluminescence, the equivalence ratio, ϕ , and the static pressure, P :

$$I \propto \dot{m}_T \phi^{5.23} P^{-0.86}, \quad (5)$$

where $\dot{m}_T = \rho U_L S$, and S is the total flame area. In Higgins' experiment, the mass flow rate but not the flame area was kept constant as the pressure was varied. However, it is easy to re-write (5) as a correlation for the OH* intensity per unit flame surface area. We can then use the pressure dependence of (5) along with the experimental temperature dependence from fig.5b to obtain a purely experimental correlation for the dependence of OH* emission per unit surface on adiabatic pressure variations. This dependence is shown by the red curve in fig. 6b. Contrary to the purely numerical results, this experimental correlation indicates that OH* chemiluminescence *increases* with pressure when the compression is adiabatic, in agreement with our observations on the response of flames to acoustic pressure waves.

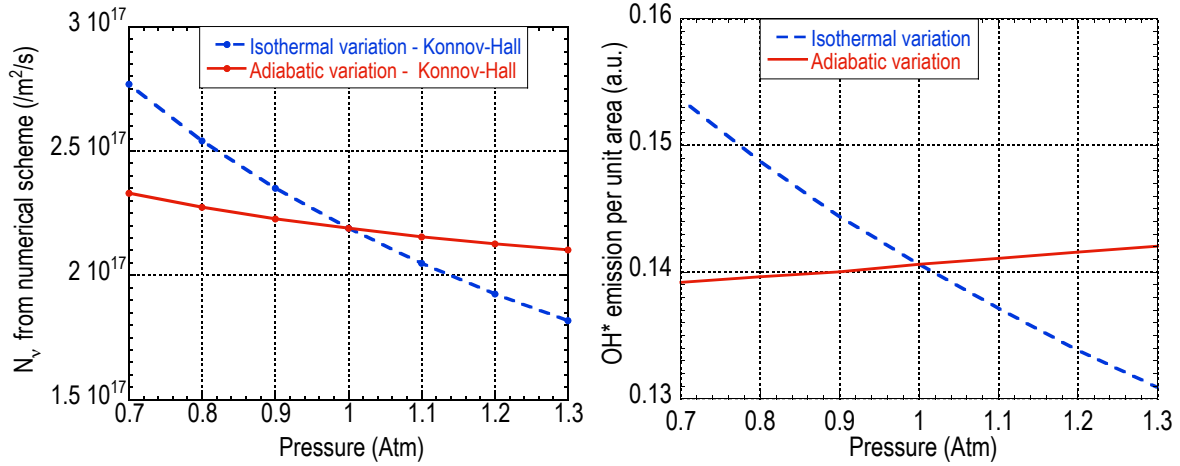


Figure 6: Pressure dependence of OH* chemiluminescence calculated with the numerical scheme of Konnov1Hall&Peterson (left) and with Higgins' experimental correlation (right).

To analyse our results on unsteady flame response, we will use a general formulation for OH* intensity

$$I = A \rho U_L f(\phi) g(T_u) h(P). \quad (6)$$

A is a constant, $\rho(T,P) U_L(\phi,T,P) = \dot{m}$ is the mass flow rate through the flame, and the three functions f , g and h describe the variations of OH* emission not directly related to changes in the mass consumption rate. Since acoustic fluctuations are very small compared to the static values, we may assume, without loss of generality, that these three functions have a power law dependence:

$$f(\phi) = \phi^x \quad ; \quad g(T) = T^y \quad ; \quad h(P) = P^z.$$

For small fluctuations of the parameters, the relative change in OH* chemiluminescence is then given by:

$$\frac{\delta I}{I} = \frac{\delta \dot{m}}{\dot{m}} + x \frac{\delta \phi}{\phi} + y \frac{\delta T_u}{T_u} + z \frac{\delta P}{P}. \quad (7)$$

Our experimental measurements of OH* intensity as functions of ϕ and T_u (fig.5) yield respectively, $x = 4.6 \pm 0.1$ and $y = 0.85 \pm 0.15$. The exponent z for the pure pressure response can then be obtained either from the experimental correlation of Higgins ($z = -0.86$) or from the numerical simulation with the kinetic scheme of Konnov+Hall&Peterson, ($z = -1.28$). These two values will lead to different results.

In the experimental measurement of the response of flames to acoustic pressure waves, the equivalence ratio is fixed, $\delta \phi = 0$, and the pressure and temperature in the fresh gas are related by the isentropic condition, equ.(2). The normalized fluctuations can then be written:

$$\frac{\delta I/I}{\delta P/P} = \frac{\delta \dot{m}/\dot{m}}{\delta P/P} + \left(\frac{(\gamma-1)}{\gamma} y + z \right) \quad (8)$$

Using our value $y = 0.85$ for the temperature exponent, taking into account that the reduced pressure fluctuations in fig.1 are normalized by $\rho c^2 = \gamma P$, we obtain

$$\frac{\delta \dot{m}/\dot{m}}{\delta P/\rho c^2} = \gamma \frac{\delta I/I}{\delta P/P} + 0.86 \text{ (Expt.) or } + 1.45 \text{ (Numeric)}, \quad (9)$$

depending on the value retained for the pressure exponent, $z = -0.86$ (experimental value) or $z = -1.28$ (value from numerical simulation). The correction, equ.(9), has been obtained for steady lean

flames in which the heat release rate is directly proportional to the mass flux through the flame, so we can write:

$$\frac{\delta \dot{Q}/\bar{\dot{Q}}}{\delta P/\rho c^2} = \gamma \frac{\delta I/I}{\delta P/P} + 0.86 \text{ (Expt.) or } +1.45 \text{ (Numeric)} \quad (10)$$

We now make the unjustified assumption that the steady state correction (10) remains valid for unsteady lean flames and apply it to our measurements of unsteady flame response. Since the physical origin of the correction arises from collision quenching of the OH* radical, and since the time scale of quenching ($\approx 10^{-12}$ s) is very much shorter than the acoustic time scale ($\approx 10^{-4}$ s), the assumption is not unreasonable. It is obvious that this correction increases the discrepancy between analytical and experimental results.

In figure 7 we have re-plotted the experimental results for flame response with the experimentally determined correction (+0.86) for the relation between OH* chemiluminescence and heat release. The analytical models now seem to under predict the flame response by at least a factor 2. The two step model with radical reaction frozen in the preheat zone (model 2) is generally closer to the experimental results, however the general trend of the experimental results is a decrease in response as the frequency increases, which is better represented by the two-step model 1, so it not possible to be conclusive.

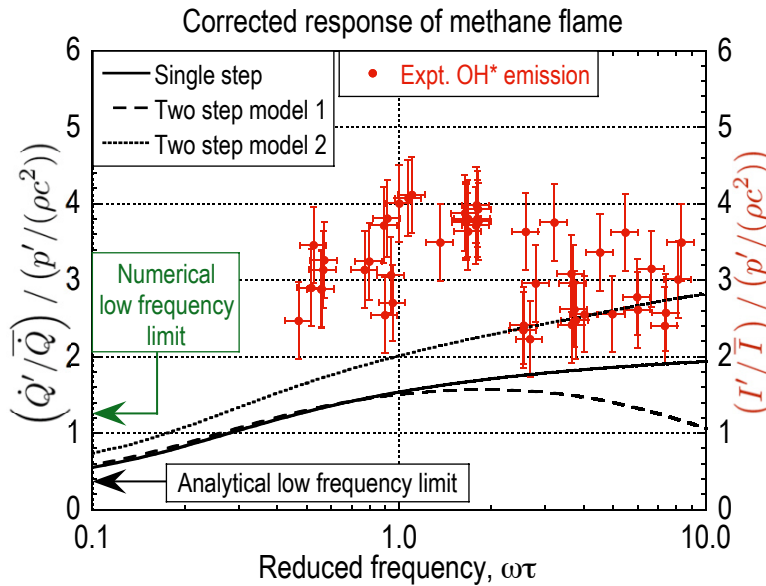


Figure 7: Comparison between corrected values of the experimental and theoretical reduced heat release rate. Experimental results are corrected with a shift of +0.86. The analytical low frequency limit and the steady state response from numerical simulations with adiabatic compression are shown by black and green arrows respectively.

The zero-frequency limit of the one-step analytical model is:

$$\frac{\dot{Q}'/\bar{\dot{Q}}}{p'/\rho c^2} = \frac{E_A}{RT_b} \frac{(\gamma-1)}{2} \frac{T_u}{T_b} \quad (11)$$

For our lean methane flames, expression (11) evaluates to 0.36. The black arrow in figure 7 shows this analytical low frequency limit. The two-step models do not have such a simple analytical expression, but their numerical evaluation yields a very similar value for the low frequency limit.

Since the kinetic mechanism of Konnov reproduces our laminar flame speeds quite well, we may expect that this scheme will also give a reasonably good prediction of the heat release response of

flames to pressure waves in the low frequency, steady state limit. We have evaluated the integrated heat release for steady flames at different pressures with $P^{1-\gamma}T^\gamma$ maintained constant (adiabatic compression). The resulting flame response, $(\dot{Q}/\ddot{Q})/(p'/\rho c^2) \approx 1.25$ is shown by the green arrow in figure 7. This value from numerical simulation is considerably higher than the value from the analytical model and lends weight to the conclusion that the analytical models substantially underestimate the unsteady response of flames to acoustic pressure waves.

CONCLUSION

We have revisited our existing experimental results for the response of premixed flames to acoustic pressure oscillations in the light of complementary experimental and numerical work relating OH* chemiluminescence to the mass consumption rate and the heat release rate. We find that pressure quenching causes the fluctuations of OH* chemiluminescence to be weaker than the corresponding fluctuations of the heat release rate and deduce a correction factor for small fluctuations.

This correction factor increases the value of the direct sensitivity of planar methane flames to acoustic pressure oscillations and increases the discrepancy between experimental results and the predictions of analytical theory. It seems probable that one of the reasons for which the analytical results underestimate the flame response is that the analytical theory neglects the sensitivity of reaction rate to density compared to the sensitivity of reaction rate to temperature. However, in order to gain a better understanding of the limitations of unsteady analytical theories and the discrepancies with experimental measurements, it would be desirable to perform full unsteady numerical simulations, both with the simple reaction kinetics used in the analytical models, and also with full chemical kinetics, including a reaction mechanism for chemiluminescence.

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